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Optimization of the Hot-Pressing Regime in the Production of Eco-Friendly Fibreboards Bonded with Hydrolysis Lignin

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Abstract

This research was aimed at studying the potential of using residual lignin from acid hydrolysis as a binder in manufacturing eco-friendly, dry-process fibreboards. For that purpose, a modification of the adhesive system and hot-pressing regime was conducted. The adhesive system applied was composed of 2 % phenol-formaldehyde (PF) resin and 10 % hydrolysis lignin (based on the dry fibres). The PF resin does not only act as a binder but generally contributes to the even distribution and good retention of the main binder – hydrolysis lignin. A specific hot-pressing cycle was used. In the first stage, the pressure was 1.0 MPa, followed by an increased pressure of 4.0 MPa, and subsequent cooling. The purpose of the initial lower pressure was softening the lignin and reduction of the material moisture content. The effect of the second stage of hot-pressing on the properties of eco-friendly fibreboards was investigated. It was determined that the fibreboards produced with 2 % PF resin and 10 % hydrolysis lignin have similar physical and mechanical properties to those of the control panels, produced with 10 % PF resin at a standard hot-pressing cycle. The findings of this work demonstrate that residual hydrolysis lignin can be effectively utilized as a binder in the production of eco-friendly, dry-process fibreboards with acceptable physical and mechanical properties.

Keywords

wood-based panels, eco-friendly, dry-process fibreboards, hydrolysis lignin, hot-pressing

1 Introduction

The development of the wood-based panel industry, related mainly to the mass introduction of continuous presses [1–3] and modification of the adhesive systems [4–8], led to the reduction of press factor from 10 ÷ 15 $s \cdot mm^{-1}$ to 4÷8 $s \cdot mm^{-1}$ [8, 9]. This reflects in increased productivity of the plants and enlarged production quantities, which in 2019 reached 357.6547 million m^3 [10]. However, the issues associated with the environmental and human-health related impact of wood-based panels, mainly connected with the hazardous emission of free formaldehyde and other volatile organic compounds (VOCs) from the finished wood-based panels, still remain relevant [11–16]. At present, the wood-based plants aim to produce panels with formaldehyde emission class E1 (≤ 8 mg/100 g). In addition, specialized formaldehyde catchers (scavengers), such as urea, amino groups, sodium metabisulfite, ammonium bisulfite or natural, bio-based formaldehyde scavengers, such as tannin powder, wheat or bark flours, etc., are often used to reduce formaldehyde emission [17, 18]. However, the panels from the E0 emission class are usually characterised by an increased production costs, hence, the industrial and scientific interest is directed at sustainable production of wood-based panels having a close-to-zero formaldehyde content, reaching E0 and super E0 classes, i.e. formaldehyde emissions equivalent to the natural wood levels [19, 20]. The use of natural, bio-based adhesives as partial or complete substitutes of the traditional thermosetting resins is a perspective approach to achieve this goal [21–29]. The use of natural binders has another environmental effect, namely the utilization of residual natural resources from other industries [30–39].

Lignin is a polyaromatic macromolecule acting as the natural binder in wood [40–42]. It is also the second most

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abundant organic polymer in the world, surpassed only by cellulose [43, 44]. At present, lignin is regarded as a waste or side-product from the pulp and paper industry as well as from biomass hydrolysis to bioethanol and other chemical products [45, 46], with an estimated annual production of approximately 100 million tons worldwide [47].

Currently, the main industrial practice is to use this valuable natural resource for heat and energy purposes [48–51] and less than 2 % is utilized in valued-added applications [47], such as dispersants, reinforcement materials, and adhesives. The main difficulty in using lignin as a binder in wood-based panels is its introduction, retention and activation [52–54]. At present, a solution to this problem is sought mainly by modification of lignin [55, 56], including its enzymatic treatment [57]. Lignin contains different functional groups, i.e., methoxyl, hydroxyl, and carbonyl groups, which allow its chemical modification, applied mostly to increase its reactivity. Significant progress has been made in replacing phenol in phenol-formaldehyde resins [58–60]. All these modifications lead to increased costs of lignin-based adhesives which makes them difficult to apply in industrial conditions. Although the most common residual product is Kraft lignin, obtained by the Kraft process [61], hydrolysis lignin, derived by enzymatic hydrolysis process, contains a certain amount of cellulose. Cellulose in hydrolysis lignin increases the contact areas between lignin and wood fibres, which leads to the formation of additional hydrogen and other bonds [62–65]. The aim of the present study was to fabricate eco-friendly dry-process fibreboards using hydrolysis lignin as a main binder by modifying the adhesive system and optimising the hot-pressing regime, without any modifications of the lignin.

2 Materials and methods

2.1 Materials

The pulp used in this research was provided by Welde Bulgaria AD (Troyan, Bulgaria). It was produced by the Asplund method with Defibrator L46 refiner (Valmet, Stockholm, Sweden), and composed of the following species: 60 % beech (*Fagus sylvatica* L.), 20 % Turkish oak (*Quercus cerris* L.) and 20 % white poplar (*Populus alba* L.). The initial moisture content of the fibres was 11 %. Technical hydrolysis lignin was used as a binder. The lignin was obtained from the high temperature diluted sulphuric acid hydrolysis of sawdust and softwood and hardwood chips to sugars, which were further subjected to yeast fodder production.

Chemical analyses of technical hydrolysis lignin were carried out according to the following methods: cellulose [66]; lignin [67], ash [68]. The analysis of C, N, S and H was performed using Elemental Analyzer Euro EA 3000. The data are presented in Table 1.

2.2 Preliminary results

Previous research on the subject has shown that lignin is not appropriate to be added in a dry state [69, 70]. In this state, it is not activated successfully and has to be used in larger quantities, which leads to sedimentation of the underside of the panels and significant deterioration of their appearance and properties of the fibreboards (Fig. 1). Therefore, hydrolysis lignin should be imported in the form of a suspension.

A study, in which hydrolysis lignin in the form of suspension was used, without an auxiliary binder, was carried out. In this case, an uneven distribution of lignin was found and hence a significant variation in the properties of the panels, Fig. 2. This led to the conclusion that an auxiliary binder should be used to retain lignin before its softening, plasticization and before forming stable bonds between lignin and wood fibres.

2.3 Experimental plan

Phenol-formaldehyde (PF) resin was used as an auxiliary binder. This type of resin was chosen because of its

Table 1 Characteristics of the technical hydrolysis lignin from the diluted sulphuric acid hydrolysis plant

Lignin, %	Cellulose, %	Ash content, %	C, %	S, %	H, %	N, %
72.6	25.5	2.8	55.54	0.74	7.10	0.26

Fig. 1 Fibreboards with hydrolysis lignin added in a dry state.

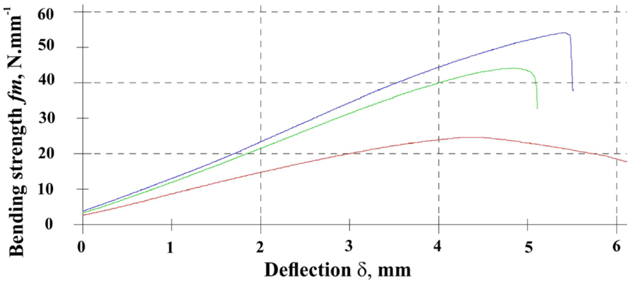

Fig. 2 Deformation behaviour under bending load of test samples from fibreboard with hydrolysis lignin without the use of an auxiliary binder.

significantly better compatibility with lignin [71–74], better adhesion properties and resistance to higher temperatures compared to urea-formaldehyde (UF) resin [75–77]. The adhesive formulation was comprised of 2 % PF resin (48.0 % dry solids content, viscosity 364 mPa s, pH 6.6, brix 72.7) manufactured by Dynea, Romania, and 10 % technical hydrolysis lignin as the main binder, based on the dry fibres. This percentage of lignin was chosen based on the previous research in the field which has shown that the most significant improvement in the properties of fibreboards is observed when the lignin content is increased to 10 % [78]. The lignin was fractionated and only the fraction below 100 μm was used.

The adhesive system, composed of hydrolysis lignin and PF resin, was introduced in the pulp at a concentration of 30 %. The introduction of the binders was carried out using a high-speed (850 min^{-1}) laboratory blender with needle-shaped blades. The adhesive composition was injected through a nozzle with a diameter of 1.5 mm, at a pressure of 0.4 MPa. Hot pressing was performed in a laboratory press Servitec - Polystat 200 T (Wustermark, Germany).

The produced panels had dimensions of 200 × 200 mm at a thickness of 4 mm and a target density of 850 $kg \cdot m^{-3}$. The panels were manufactured at a hot-pressing temperature of 200 °C. The press temperature was chosen based on the results of previous studies, which confirmed the improvement of the fibreboard properties with increasing temperature to this range [79, 80].

The panels with hydrolysis lignin were produced using a modified hot-pressing cycle: pressure in the first stage was 1.0 MPa (low pressure), pressure in the second stage was 4.0 MPa (high pressure) and subsequent cooling while maintaining the high pressure. The panels with hydrolysis lignin were produced using a modified hot-pressing cycle: pressure in the first stage was 1.0 MPa (low pressure), pressure in the second stage was 4.0 MPa (high pressure) and subsequent cooling while maintaining the high pressure. The value of the pressure in the first stage was chosen given the possibility of removing the vapour from the material, and in the second stage because of its densification. These pressure values are in line with those commonly used in the industry for that purpose [1, 81].

The pressing time of the first stage was constant - 5 min. This time was determined experimentally. For the conditions of the experiment (11 % moisture content of the fibres and 30 % concentration of the adhesive system) in a shorter time, the vapour cannot be separated from the material, as a result of which combustion and hydrolysis occur in the components of the panels.

The press factor of the second stage varied as follows:

- Panel type A – 7.5 $s \cdot mm^{-1}$ (pressing time of 30 s),
- Panel type B – 15 $s \cdot mm^{-1}$ (pressing time of 60 s),
- Panel type C – 22.5 $s \cdot mm^{-1}$ (pressing time of 90 s),
- Panel type D – 30 $s \cdot mm^{-1}$ (pressing time of 120 s) and
- Panel type E – 90 $s \cdot mm^{-1}$ (pressing time of 360 s).

The purpose of this modified cycle was to plasticize and soften the lignin during the first low-pressure stage. Similar studies have shown that the plasticization of lignin leads to significantly better properties of the fibreboards [82]. Another modification of the pressing regime, namely cooling at the end of the hot-pressing, was applied. The cooling was aimed at converting the lignin, which has already formed bonds with the fibres, to a solid state. The cooling time was 3 minutes (until the temperature reached values below 100 °C). Reference panels, with 10 % PF resin (REF 10) and a standard hot-pressing cycle were also produced - the first stage 4.0 MPa; second stage 1.2 MPa; third stage 0.6 MPa and fourth stage 1.5 MPa. Control panels were produced at a press-factor of 30 $s \cdot mm^{-1}$. The press temperature used was 200 °C. Following the hot pressing, the fibreboards were conditioned for seven days at 20 ± 2 °C and 65 % relative humidity.

From each type of boards were produced three panels. The physical and mechanical properties of the fibreboards were determined by testing 8 test samples for each property. The properties of the panels were determined according to the methods defined in the EN [83–85]. A Zwick / Roell Z010, Ulm, Germany universal testing machine was used for the determination of the mechanical properties of the panels.

3 Results and analyses

The laboratory-produced panels are presented in Fig. 3. The density of the panels is presented in Table 2.

The manufactured panels had a density rather close to the targeted value of 850 $kg \cdot m^{-3}$. The highest density was determined for Panel Type D – 876 $kg \cdot m^{-3}$, and the lowest

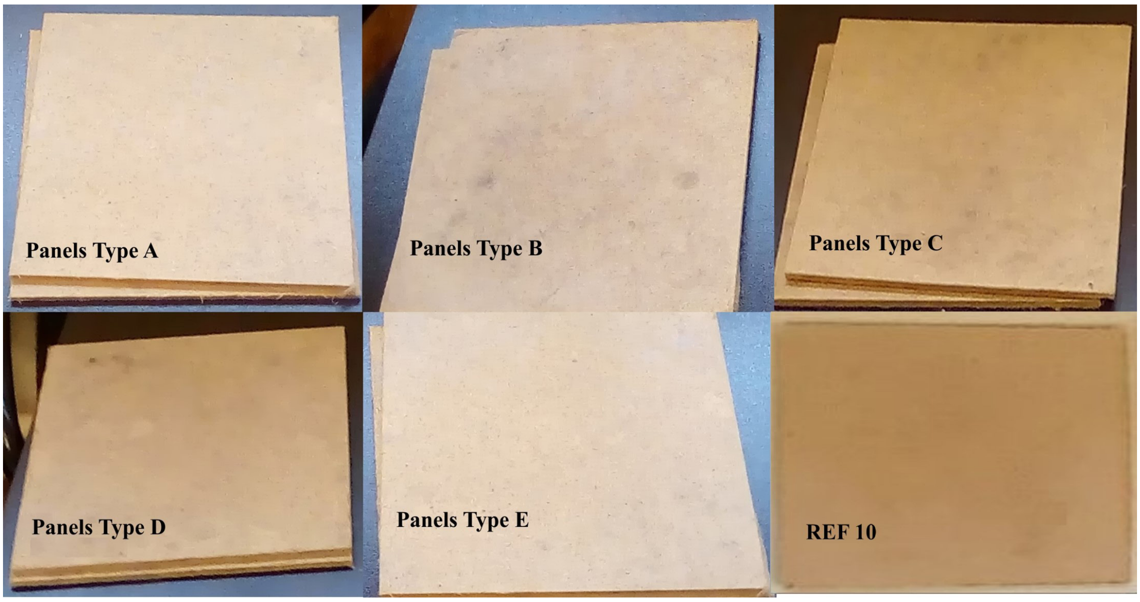

Fig. 3 Eco-friendly dry-process fibreboard panels with an adhesive system comprised of 10 % hydrolyses lignin and 2 % PF resin. Type A – second stage press factor 7.5 s·mm^{-1}; Type B – second stage press factor 15 s·mm^{-1}; Type C – second stage press factor 22.5 s·mm^{-1}; Type D – second stage press factor 30 s·mm^{-1}; Type E – second stage press factor 90 s·mm^{-1}; REF 10 – 10 % PF resin.

Table 2 Density of the eco-friendly dry-process fibreboard panels with an adhesive system comprised of 10 % hydrolysis lignin and 2 % PF resin

Panel Type	Type A	Type B	Type C	Type D	Type E	REF 10
Density ρ, kg.m^{-3}	861 ± 20	865 ± 17	867 ± 15	876 ± 22	854 ± 12	871 ± 6

for Panel Type E – 854 kg·m^{-3}. The difference between these two density values was 2.58 %, i.e. significantly below the permissible statistical error of 5 %. The p-value, after conducting a t-test for the densities of these two panels is 0.258 (above 0.05), i.e. there was no statistical difference between the density mean values of the panels. Thus, it can be concluded that the density of the panels did not have a significant effect on the other physical and mechanical properties of the panels, and the deviation of the properties will result from altering the hot-pressing regime.

The results for the water absorption (WA) of the panels are presented in Fig. 4.

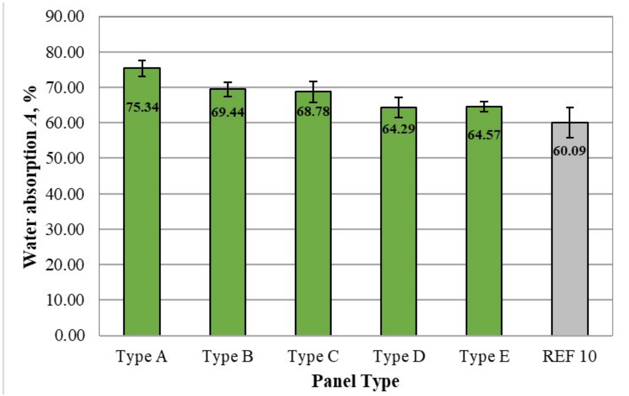

Fig. 4 Water absorption (24 h) of eco-friendly dry-process fibreboards bonded with 10 % hydrolysis lignin and 2 % PF resin. Type A – second stage press factor 7.5 s·mm^{-1}; Type B – second stage press factor 15 s·mm^{-1}; Type C – second stage press factor 22.5 s·mm^{-1}; Type D – second stage press factor 30 s·mm^{-1}; Type E – second stage press factor 90 s·mm^{-1}; REF 10 – 10 % PF resin.

WA of the panels with an adhesive system of 2 % PF resin and 10 % hydrolysis lignin varied from 75.34 % to 64.29 %. The main decrease (improvement) in the value of the property by 1.08 times was determined when the second stage press factor was increased from 7.5 to 15 s·mm^{-1}. The next improvement resulted from the increase of press factor from 22.5 to 30 s·mm^{-1}. In that case, the WA of the panels was improved by 1.07 times. However, the conducted t-tests showed that the only statistical difference was between the WA of the panels produced with a press factor of 7.5 s·mm^{-1} and those at 15 s·mm^{-1}. The p-value from this t-test was 0.009. This revealed that regarding the WA it is not justified to extend the second stage press factor above 15 s·mm^{-1}.

A graphical representation of the thickness swelling (TS) values (24 h) of the laboratory-produced panels is presented in Fig. 5.

The TS of the panels produced with hydrolysis lignin as the main binder varied from 34.37 % to 27.53 %. Therefore, the increase of the second stage press factor led to an improvement of that property by 1.25 times. The main improvement was determined at the increase of

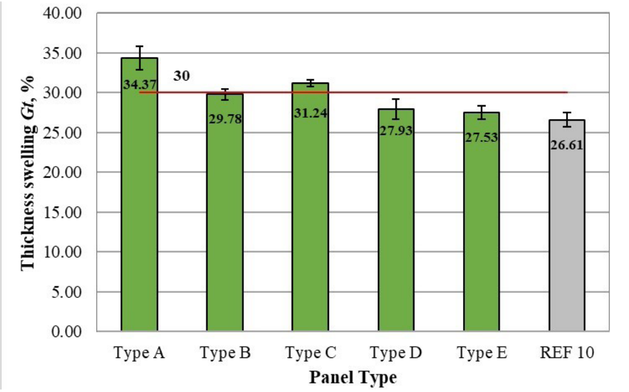

Fig. 5 Thickness swelling (24 h) of eco-friendly dry-process fibreboards bonded with 10 % hydrolysis lignin and 2 % PF resin (the red line represents the EN 622-5 standard requirement for load-bearing boards for use in humid conditions). Type A – second stage press factor 7.5 s·mm^{-1}; Type B – second stage press factor 15 s·mm^{-1}; Type C – second stage press factor 22.5 s·mm^{-1}; Type D – second stage press factor 30 s·mm^{-1}; Type E – second stage press factor 90 s·mm^{-1}; REF 10 – 10 % PF resin.

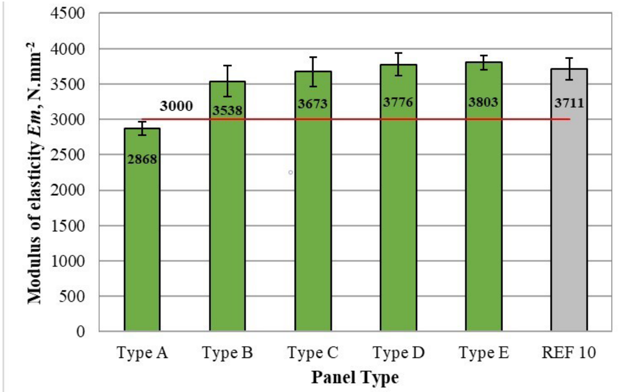

Fig. 6 Modulus of elasticity (MOE) of eco-friendly dry-process fibreboards bonded with 10 % hydrolysis lignin and 2 % PF resin (the red line represents the EN 622-5 standard requirement for load-bearing applications for use in humid conditions). Type A – second stage press factor 7.5 s·mm^{-1}; Type B – second stage press factor 15 s·mm^{-1}; Type C – second stage press factor 22.5 s·mm^{-1}; Type D – second stage press factor 30 s·mm^{-1}; Type E – second stage press factor 90 s·mm^{-1}; REF 10 – 10 % PF resin.

press factor from 7.5 s·mm^{-1} to 15 s·mm^{-1}. In this case, the improvement was by 1.15 times.

With t-tests, it was established that there was no statistical difference between the thickness swelling of the panels produced at press factors 15 and 22.5 s·mm^{-1} (p-value is 0.17). A significant improvement in the TS was observed when the second stage press factor was increased from 22.5 to 30 s·mm^{-1}. The improvement is 1.12 times. Again, after a t-test, it was found that there was no statistical difference between the values for swelling in the thickness of the panels at a press factor of 30 and 90 s·mm^{-1} (p-value is 0.20). All this leads to the conclusion that for the improvement of the TS values of eco-friendly fibreboards bonded with hydrolysis lignin, it is not justified the press factor of the second stage to be above 30 s·mm^{-1}.

Markedly, the panels manufactured with 2 % PF resin and 10 % hydrolysis lignin at modified hot-pressing cycle and second stage press factor of 30 s·mm^{-1}, had TS values similar to that of the panels with 10 % PF resin, produced at standard hot-pressing cycle. The performed t-test showed that there was no statistical difference between these two types of panels – the p-value is 0.27.

All panels with hydrolysis lignin as the main binder met the standard requirements for TS for load-bearing boards and use in dry conditions. Panels with a press factor of 15 s·mm^{-1} met the most stringent requirements for TS, namely for load-bearing applications for use in humid conditions [86].

The results for the modulus of elasticity (MOE) of the panels are given in Fig. 6.

The laboratory-produced fibreboards, bonded with hydrolysis lignin exhibited MOE values, ranging from 2868 to 3803 N·mm^{-2}. That is, the increase in the second stage press factor above 7.5 s·mm^{-1}, resulted in improvement of the property by 32.60 %. It should be noted that the only statistical difference between the properties for the individual panels was determined for the fibreboards produced with a press factor of 7.5 s·mm^{-1} and those with a press factor of 15 s·mm^{-1} (p-value is 0.01). Further increase in the press factor did not lead to a significant improvement of the MOE values. After performed the t-test, it was found that there was no statistical difference between the MOE of the panels produced at a press factor of 15 s·mm^{-1} and the reference panels with 10 % PF resin (p-value is 0.42). Therefore, eco-friendly fibreboards with MOE values, similar to those of fibreboards produced with 10 % PF resin, can be manufactured by using an adhesive composition of only 2 % PF resin and 10 % hydrolysis lignin, with a second stage press factor of 15 s·mm^{-1}.

All laboratory-fabricated panels bonded with hydrolysis lignin met the requirements regarding the MOE of fibreboards for general purpose and use in humid conditions. Fibreboards produced at the second stage press factor of 15 s·mm^{-1} met the strictest requirements for the property for load-bearing boards and use in humid conditions [86].

The results for the bending strength (MOR) of the panels are presented in Fig. 7.

For the experimental conditions, the MOR values of eco-friendly fibreboards bonded with 10 % hydrolysis lignin varied from 31.24 N·mm^{-2} to 43.69 N·mm^{-2}. That is,

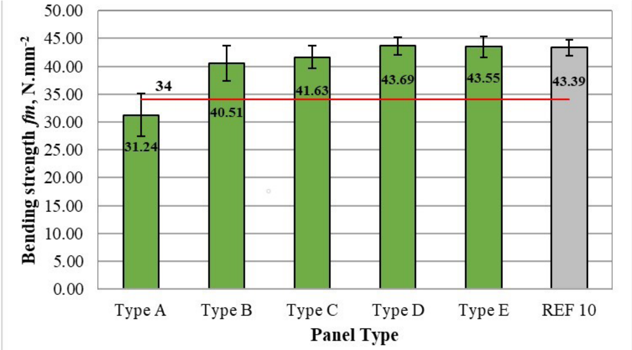

Fig. 7 Bending strength (MOR) of eco-friendly dry-process fibreboards bonded with 10 % hydrolysis lignin and 2 % PF resin (the red line represents the EN 622-5 standard requirement for load-bearing applications for use in humid conditions). Type A – second stage press factor 7.5 s·mm^{-1}; Type B – second stage press factor 15 s·mm^{-1}; Type C – second stage press factor 22.5 s·mm^{-1}; Type D – second stage press factor 30 s·mm^{-1}; Type E – second stage press factor 90 s·mm^{-1}; REF 10 – 10 % PF resin.

the improvement in this property was by 39.85 %. It should be emphasized that the only statistical difference was in the panels produced at the second stage press factor of 7.5 s·mm^{-1} and those at 15 s·mm^{-1} (p-value is 0.005). That is, it was confirmed that given the mechanical properties of the panels, the second stage press factor of 7.5 s·mm^{-1} is insufficient for the formation of stable bonds between the hydrolysis lignin and the wood fibres. However, at a press factor of 15 s·mm^{-1}, panels with very good mechanical properties could be produced, and a subsequent increase in the second stage press factor did not lead to further improvement. This is why the optimal second stage press factor for the production of eco-friendly fibreboards bonded with 10 % hydrolysis lignin as the main binder was 15 s·mm^{-1}. The performed t-test showed that there was no statistical difference between the MOR values of the panels produced at the second stage press factor of 15 s·mm^{-1} and the reference panels with 10 % PF resin (p-value is 0.187), i.e. the eco-friendly fibreboards with 2 % PF resin and 10 % hydrolysis lignin, produced at a second stage press factor of 15 s·mm^{-1} had a MOR values, quite comparable to those of the panels with 10 % PF resin.

All eco-friendly fibreboards bonded with 10 % hydrolysis lignin met the requirements for general-purpose fibreboards for use in humid conditions. With the exception of the panel produced at the second stage press factor of 7.5 s·mm^{-1}, all other eco-friendly panels met the strictest requirements for the property – for load-bearing boards and use in humid conditions [86].

The obtained results for eco-friendly fibreboards manufactured with a modified adhesion system and press cycle were comparable and better than those reported in similar studies [69, 79, 80, 87]. The obtained eco-friendly fibreboards had a lower density and better properties than those obtained by Zhou et al. [79], and the lignin is not enzymatically modified. Compared with the study by Tupciauskas et al. [69], the obtained fibreboards had similar properties, but at a much lower density of 850 kg·m^{-3} versus 1300 kg·m^{-3}, at a significantly lower lignin content of 10 % versus 25 %. In addition, the hydrolysis lignin used in the present study did not impair the appearance of the panels. The results obtained are also comparable to those obtained by Theng et al. [80], but again at a lower density of fibreboards and lower lignin content than in the study by Nasir et al. [87].

4 Conclusions

In the present study, it was found that hydrolysis lignin can be successfully used as a binder with very good performance in the production of eco-friendly dry-process fibreboards. A solution is proposed to eliminate the main disadvantages of hydrolysis lignin as a binder – its introduction and retention in the fibres and its activation in the process of hot-pressing. The results of the study show that the hydrolysis lignin is activated by modifying the hot-pressing cycle, without the need to apply additional modifications or treatment with enzymes. The modifications of lignin proposed in previous studies significantly increase costs and make it difficult to apply in industrial conditions. The optimization of the hot-pressing cycle is easily feasible in industrial conditions when using continuous presses, which have autonomous heating of the individual sections. That is, cooling will not lead to additional costs for reheating, which was typical of older technologies using multi-story presses.

The implemented modification of the adhesive system overcomes another major disadvantage of hydrolysis lignin, namely its retention in the pulp. The small amount of PF resin used (comparable to that used in the wet-process) performs precisely the role of lignin retention and partial auxiliary function as a binder.

The study showed that to achieve the most stringent requirements for mechanical properties of eco-friendly panels, it is not necessary to extend the second stage press factor above 15 s·mm^{-1}, and above 30 s·mm^{-1}, regarding the water-repellent properties. It is very promising that the requirements for general-purpose fibreboards (the most common type on the market) are achieved even with a second stage press factor of 7.5 s·mm^{-1}. Future studies should be focused on the optimization of the first stage of the pressing regime and establishing the optimal moisture content of the fibre mat at which hydrolysis lignin is activated.

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